

Electronic nematic correlations in the stress free tetragonal state of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$

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We use transport and neutron scattering to study electronic, structural, and magnetic properties of the electron-doped $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ iron pnictides in the external stress free detwinned state. Using a specially designed in-situ mechanical detwinning device, we demonstrate that the in-plane resistivity anisotropy observed in the uniaxial strained tetragonal state of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ below a temperature T^* , previously identified as a signature of the electronic nematic phase, is also present in the stress free tetragonal phase below T^{**} ($< T^*$). By carrying out neutron scattering measurements on BaFe_2As_2 and $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$, we argue that the resistivity anisotropy in the stress free tetragonal state of iron pnictides arises from the magnetoelastic coupling associated with antiferromagnetic order. These results thus indicate that the local lattice distortion and nematic spin correlations are responsible for the resistivity anisotropy in the tetragonal state of iron pnictides.

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There is growing experimental evidence suggesting that the electronic nematic phase, a translationally invariant metallic phase (satisfy the 90° -rotational or C_4 symmetry) with a spontaneously generated spatial electronic anisotropy, is intimately connected with high-transition (high- T_c) temperature superconductivity [1]. For iron pnictide superconductors such as $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($T = \text{Co, Ni}$) [2–7], their parent compound BaFe_2As_2 exhibits a tetragonal to orthorhombic structural phase transition at temperature T_s , followed by a paramagnetic to antiferromagnetic (AF) phase transition at T_N ($T_s \geq T_N$) with a collinear AF structure [Fig. 1(a)] [5–7]. Upon electron-doping via Co or Ni substitution for Fe, the T_N and T_s are gradually suppressed and optimal superconductivity emerges near $x \approx 0.1$ for $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ [8–12]. Due to the formation of twin domains in the orthorhombic state of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ below T_s , the intrinsic electronic properties of these materials can be probed by applying a uniaxial pressure (strain) along one-axis of the orthorhombic lattice to detwin the single crystal [13–16]. While there is indeed a large in-plane resistivity anisotropy in the uniaxial strain detwinned $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ below T_s , the anisotropy persists in the paramagnetic tetragonal state below a characteristic temperature T^* ($T^* > T_s \geq T_N$), thus suggesting the presence of electronic nematic correlations above T_s and below T^* [3, 13–17]. However, since the uniaxial strain necessary to detwin the sample also enhances T_N [19] and introduces an explicit symmetry breaking field, it is unclear if there will be resistivity anisotropy in the stress free tetragonal state below T^* upon releasing the applied external uniaxial strain. From transport [3, 16, 17], inelastic neutron scattering [20], and ther-

modynamic measurements [21], T^* is believed to mark a temperature range of nematic fluctuations with structure and magnetic phase transitions occurring at T_s and T_N , respectively. On the other hand, magnetic torque and X-ray diffraction experiments on stress free samples of BaFe_2As_2 suggest that T^* is a signature of a “true” second-order nematic phase transition from the high-temperature tetragonal phase to a low-energy orthorhombic phase [22]. To understand the role of electronic nematic phase in high- T_c superconductivity, it is important to reveal the origin of the resistivity anisotropy above T_s without external uniaxial strain and determine the nature of the nematic correlations below T^* [23].

We use transport and neutron scattering to study the resistivity anisotropy, magnetic order, and lattice distortion in parent compound BaFe_2As_2 ($T_N \approx T_s \approx 138$ K) and electron-doped $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0.015, 0.03, 0.05, 0.065$). In previous transport and neutron scattering measurements, the applied uniaxial pressure necessary to detwin the crystal in the orthorhombic AF phase remains in the paramagnetic tetragonal state ($T > T_N, T_s$), thus complicating the interpretation of the observed in-plane resistivity and spin excitation anisotropy [3, 13, 14, 16, 17, 19, 20]. To avoid this problem, we have designed an in-situ mechanical sample clamp which can apply and release uniaxial pressure at any temperature, similar to the device used to study the anisotropic optical response in iron pnictides [24]. Figure 1(b) shows the schematics of the sample stick with a micrometer on the top. The magnitude of the uniaxial pressure along the b -axis direction of the orthorhombic lattice is applied by a spring that is controlled by the displacement of the micrometer (and external applied pressure) [Fig. 1(c)]. By applying uniaxial pressure at

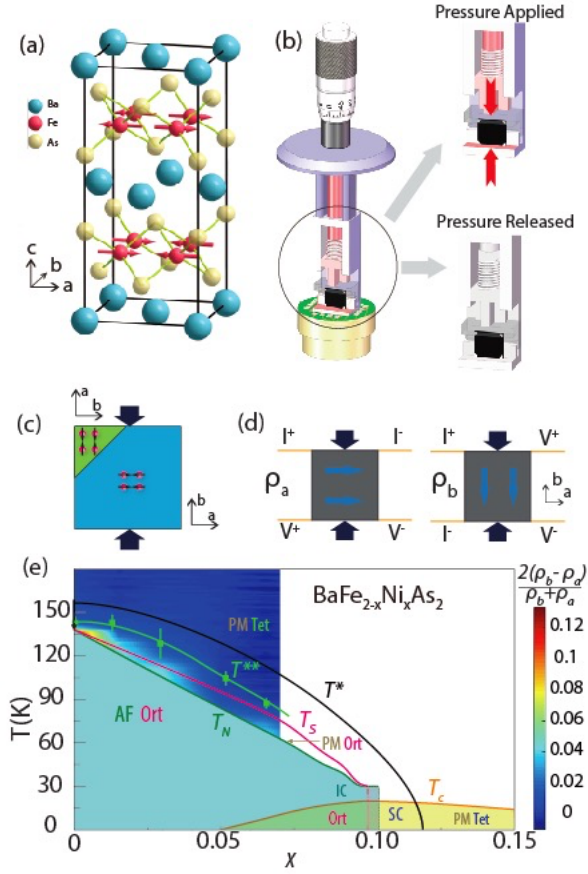


FIG. 1: Summary of transport and neutron scattering results. (a) The crystal and magnetic structures of BaFe₂As₂ in the AF orthorhombic state where the arrows mark the moment directions of iron [6]. (b) Schematic diagram of the device used to change pressure on the sample in-situ. A micrometer is used to adjust the length of the spring and therefore the pressure applied on the sample. The applied pressure then can be released by fully retreat of the micrometer, as indicated in the expanded schematic on the right. (c) The uniaxial strain is applied along the *b*-axis of the crystal, enlarging the blue domain and reducing the green domain. (d) Wire connection and current flow directions for resistivity measurements using Montgomery method. The black arrows indicate the uniaxial pressure direction and the blue arrows in the sample are the current direction for each set up. (e) The electronic phase diagram of BaFe_{2-x}Ni_xAs₂ as a function of Ni-doping as determined from previous experiments [12]. The spin excitation anisotropy temperatures under uniaxial strain are marked as T^* [20]. The AF orthorhombic (Ort), incommensurate AF (IC) [12], paramagnetic tetragonal (PM Tet), superconductivity (SC) phases are clearly marked. T^{**} marks the temperature below which resistivity anisotropy appears in the strain free tetragonal state.

room temperature (above T_N and T_s), cooling the sample to below T_N , and then releasing the pressure, we can in principle obtain the single domain sample without external strain (stress free). To conclusively determine the sample detwinning ratio and compare them with the re-

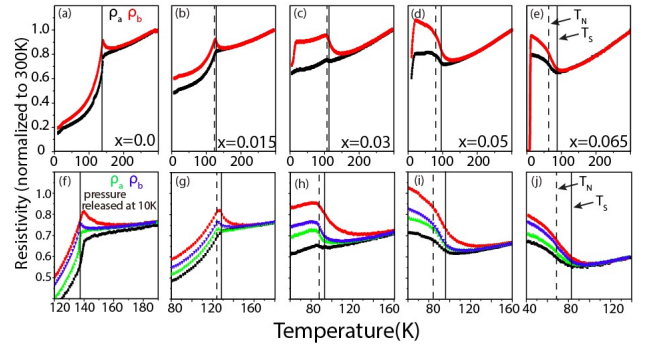


FIG. 2: Temperature dependence of the resistivity anisotropy in strained and strain free BaFe_{2-x}Ni_xAs₂. (a-e) Temperature dependence of the in-plane resistivity ρ_a (black) and ρ_b (red) under uniaxial strain for BaFe_{2-x}Ni_xAs₂ with $x = 0, 0.015, 0.03, 0.05, 0.065$, respectively. The vertical solid and dashed lines mark T_s and T_N , respectively, for these materials without uniaxial strain. (f-j) Expanded view of the data in (a-e). The green and blue data points are ρ_a and ρ_b resistivity obtained on warming after releasing the pressure at 10 K. In all cases, the resistivity is measured on heating, with the same sample and same contacts (four point Montgomery method).

sistivity anisotropy measurement, we used two original sample sticks one for transport in a physical property measurement system (PPMS) and one for neutron scattering experiment on IN8 triple-axis spectrometer at Institut Laue-Langevin (ILL). Our key finding is that the resistivity anisotropy in BaFe_{2-x}Ni_xAs₂ seen in the uniaxial strained tetragonal phase below T^* is also present in the stress free tetragonal state, but at a lower temperature $T^{**} < T^*$ [Fig. 1(e)]. In addition, our neutron Larmor diffraction measurements [25, 26] on temperature dependence of the lattice spacing (*d*) and its distortion (Δd) in lightly electron-doped iron pnictides reveal that the lattice distortion increases on cooling, passes smoothly across T_s , and enhances dramatically on approaching T_N with no observable anomaly above T_s . These results suggest that the resistivity anisotropy observed in the external uniaxial pressure free tetragonal state of BaFe_{2-x}Ni_xAs₂ arises from a strong magnetoelastic coupling induced by AF order, and there are no additional thermodynamic phase transitions above T_s [21, 27]. Therefore, the Ising-nematic correlations, a state with no magnetic long-range order (staggered magnetization $M = 0$) but with local broken C_4 symmetry lattice distortion [28–30], is the driving force for the observed resistivity anisotropy [23, 28–30].

We first compare transport measurements obtained on single domain samples detwinned using a standard mechanical clamp and the new device [Fig. 1(b)]. The resistivity data along the orthorhombic *a* and *b* directions are measured via the Montgomery method [31]. Resistivity along the *a* (ρ_a) and *b* (ρ_b) directions are measured

in the same cycle using different current directions with wiring diagram shown in Fig. 1(d). Two sets of resistivity data as a function of temperature were collected for the detwinned crystals of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$. Figure 2(a)-2(e) shows temperature dependence of ρ_a and ρ_b for $x = 0, 0.015, 0.03, 0.05, 0.065$, respectively, under ~ 10 MPa of uniaxial pressure. Consistent with previous work [15], we see clear resistivity anisotropy ($\rho_b > \rho_a$) at temperatures above the strain free T_N and T_s marked as vertical dashed and solid lines, respectively. The green and blue lines in Figures 2(f)-2(j) show ρ_a and ρ_b , respectively, on the warming cycle when the room-temperature applied strain is released at base temperature (10 K). The corresponding ρ_a and ρ_b under uniaxial strain in Fig. 2(a)-2(e) are shown as black and red lines in Fig. 2(f)-2(j).

In the undoped parent compound ($x = 0$), the uniaxial strain clearly increases the temperature below which the resistivity decreases with decreasing temperature [Fig. 2(f)], consistent with the notion that the uniaxial strain necessary for detwinning the sample also increases the T_N of the system [19]. In addition, we see that the uniaxial strain itself enhances the resistivity anisotropy both below and above T_N (T_s). Although much reduced, the resistivity anisotropy ($\rho_b > \rho_a$) is also present in the stress free tetragonal state above T_N (T_s). When the Ni-doping level is increased to $x = 0.015, 0.03$, we find similar trend for strained and stress free resistivity [Figs. 2(g) and 2(h)]. Since T_N and T_s are now clearly separated, we can see that the resistivity reduction in the stress free sample happens below T_N , and the resistivity anisotropy shows no observable anomaly across T_s . Upon further increasing the Ni-doping levels to $x = 0.05, 0.065$, the resistivity smoothly increases on cooling across T_s and no longer displays a clear kink below T_N . At all doping levels studied, we find resistivity anisotropy in stress free samples above T_N and T_s (Fig. 2).

Although transport data in Fig. 2 revealed clear evidence for resistivity anisotropy in the stress free tetragonal state of underdoped $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ [Fig. 1(e)], these measurements cannot determine the sample detwinning ratio upon releasing the uniaxial strain at low temperature and microscopic origin of the resistivity anisotropy above T_s . To address these questions, we carried out neutron diffraction experiments on BaFe_2As_2 using an in-situ detwinning device similar to Fig. 1(b) and the flat-cone option of the IN8 triple-axis spectrometer at ILL [32]. In addition, we performed neutron Larmor diffraction measurement on $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ using the TRISP triple-axis at Heinz Maier-Leibnitz, Garching, Germany [25, 26]. We first describe neutron diffraction experiments on IN8 designed to study the detwinning ratio and its temperature dependence in strained and stress free BaFe_2As_2 , as these results will allow us to determine if the detwinning ratio is maintained after releasing the uniaxial strain below T_N . For the experiment,

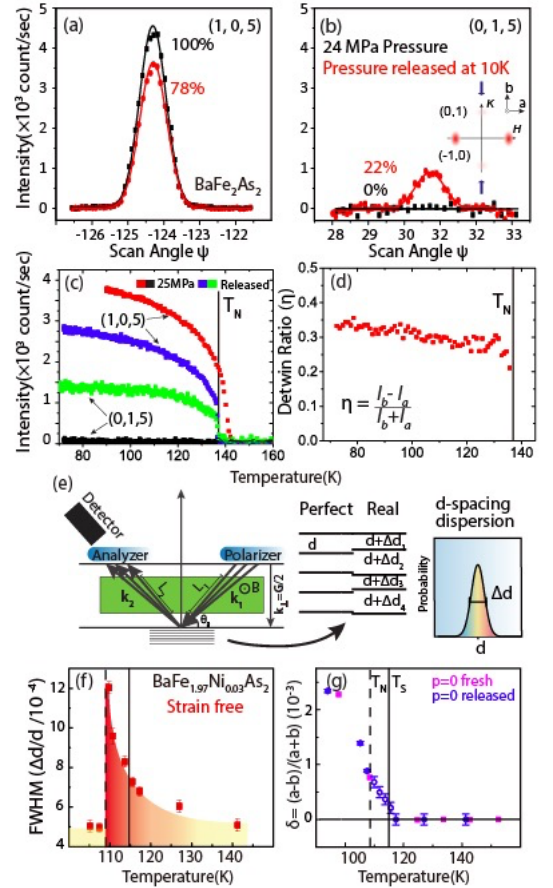


FIG. 3: Temperature dependence of Magnetic Bragg peaks at (1, 0, 5) and (0, 1, 5) in strained and strain free BaFe_2As_2 and neutron Larmor diffraction in $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$. (a) Transverse scan through magnetic Bragg peak (1, 0, 5) with and without uniaxial strain at 10 K obtained using flatcone setup and in-situ sample clamp on IN8. (b) Identical scans through (0, 1, 5) peak on IN8. (c) Temperature dependence of the magnetic scattering at (1, 0, 5) and (0, 1, 5) in strained and stress free case. (d) Estimated temperature dependence of the detwinning ratio η . (e) Schematic diagram for the configuration of neutron Larmor diffraction measurements [25, 26]. In neutron Larmor diffraction, the neutron precession directions are same in L_1 and L_2 . It can accurately measure lattice spacing d and its distortion Δd . (f) Temperature dependence of the $\Delta d/d$. The solid and dashed vertical lines are T_s and T_N , respectively. (g) Temperature dependence of the estimated orthorhombicity δ for fresh and strain free (first apply uniaxial pressure, then release pressure) $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$. The most dramatic changes in lattice distortion happen at T_N and not at T_s .

an annealed square-shaped single crystal of BaFe_2As_2 (~ 220 mg) was mounted on a specially designed sample stick inside an orange cryostat. The momentum transfer \mathbf{Q} in three-dimensional reciprocal space in \AA^{-1} is defined as $\mathbf{Q} = H\mathbf{a}^* + K\mathbf{b}^* + L\mathbf{c}^*$, where H , K , and L are Miller Indices and $\mathbf{a}^* = \hat{\mathbf{a}}2\pi/a$, $\mathbf{b}^* = \hat{\mathbf{b}}2\pi/b$, $\mathbf{c}^* = \hat{\mathbf{c}}2\pi/c$ [12]. In the AF ordered state of a 100%

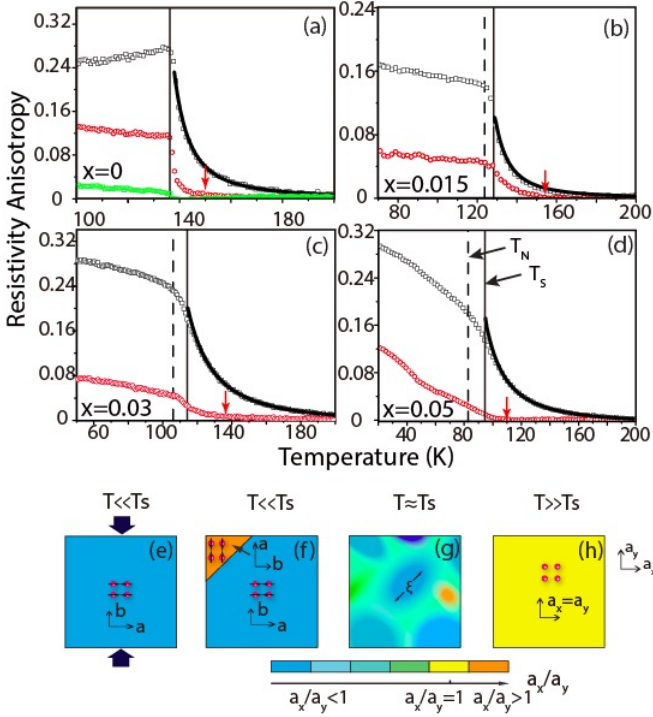


FIG. 4: Temperature dependence of the in-plane resistivity anisotropy, defined as $2(\rho_b - \rho_a)/(\rho_b + \rho_a)$, for strained and strain free $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$. (a-d) Temperature dependence of the in-plane resistivity anisotropy for $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ under uniaxial pressure (Black) and pressure released at 10 K (Red) on warming. Solid and dashed vertical lines mark T_s and T_N , respectively, for each Ni-doping. The solid black line is a fit above T_s using Curie-Weiss functional form. The red arrows mark the estimated T^{**} . The green curve in (a) represents resistivity anisotropy measured on cooling in stress free case from room temperature. (e-h) Microscopic picture of what happens in the process of releasing uniaxial strain at low temperature. a and b are orthorhombic lattice parameters. At temperatures slightly above T_s in strain free case, the overall crystal structure is tetragonal but there are local orthorhombic lattice distortions induced by the strong magnetoelastic coupling, which gives rise to the observed resistivity anisotropy.

detwinned sample, the AF Bragg peaks should occur at $(\pm 1, 0, L)$ ($L = 1, 3, 5, \dots$) positions in reciprocal space and be absent at $(0, \pm 1, L)$. Our sample is aligned in the $[H, 0, L]$ scattering plane. Using the flatcone setup on IN8 [32], we can access both $(1, 0, 5)$ and $(0, 1, 5)$ Bragg positions. When a pressure of ~ 24 MPa is applied along the b direction of BaFe_2As_2 , the sample is 100% detwinned with no magnetic scattering at $(0, 1, 5)$ [Fig. 3(a) and 3(b)]. After releasing the uniaxial pressure at 10 K, we see that the sample becomes partially twinned again with magnetic scattering intensity at both $I(1, 0, 5)$ and $I(0, 1, 5)$, giving a detwinning ratio of $\eta \approx 56\%$ ($\approx [I(1, 0, 5) - I(0, 1, 5)]/[I(1, 0, 5) + I(0, 1, 5)]$). This is consistent with transport measurements indicating a smaller resistivity anisotropy in the stress free BaFe_2As_2

[Fig. 2(f)]. Figure 3(c) shows temperature dependence of the magnetic scattering at $(1, 0, 5)$ and $(0, 1, 5)$ under 25 MPa uniaxial pressure and stress free. While the sample is 100% detwinned under 25 MPa below T_N with no magnetic scattering at $(0, 1, 5)$, the stress free sample has finite intensity at both $(1, 0, 5)$ and $(0, 1, 5)$ below T_N . Figure 3(d) shows temperature dependence of η , which reveals a decreasing detwinning ratio on warming to T_N .

In previous studies of the neutron extinction effect on the $(2, -2, 0)$ nuclear Bragg peak of BaFe_2As_2 in zero pressure [20], its intensity is found to deviate from normal behavior below ~ 150 K before displaying a step like feature at $T_N \approx T_s \approx 138$ K, suggesting the presence of fluctuating orthorhombic structural domains above T_s . Using neutron Larmor diffraction with polarized neutrons [Fig. 3(e)], we can precisely determine temperature dependence of the lattice parameter and its distortion [25, 26]. Since transport measurements in Fig. 2 suggest that the resistivity anisotropy in stress free detwinned sample reduces dramatically above T_N and shows no visible anomaly across T_s for lightly electron-doped $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ [Fig. 2(g) and 2(h)], we decided to study temperature dependence of the lattice distortions and orthorhombicity $\delta = (a - b)/(a + b)$ in $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ [7], where T_s and T_N are well separated as determined from transport and neutron diffraction experiments. For this purpose, we focus on $(4, 0, 0)$ Bragg peak, which has a d -spacing $d = a/4$. In a classical second order magnetic phase transition, one would expect that spin-spin correlation length increases on cooling and diverges at T_N , while the underlying lattice correlations ξ remain long-ranged and temperature independent. Surprisingly, our neutron Larmor diffraction measurements on stress free $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ reveal that the lattice distortion $(\Delta d/d)$ of the system shows no visible anomaly across T_s (≈ 118 K), but increases continuously on cooling below T_s before collapsing abruptly below T_N (≈ 109 K) [Fig. 3(f)] [33]. Similarly, instead of being a temperature independent constant, the lattice correlation length ξ decreases on cooling, changing smoothly from 2500 Å around ~ 150 K to 1000 Å just above T_N with no anomaly across T_s [33]. Figure 3(g) compares temperature dependence of the lattice orthorhombicity δ for $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ without applying any external strain and in strain released sample. In both cases, we see that AF order induces a large change in lattice orthorhombicity, consistent with previous X-ray scattering work [7]. Therefore, $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ exhibits a strong magnetoelastic coupling near T_N .

Figure 4 summarizes temperature dependence of the resistivity anisotropy, defined as $2(\rho_b - \rho_a)/(\rho_b + \rho_a)$, for uniaxial strained and stress free $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ with $x = 0, 0.015, 0.03, 0.05$. Similar to previous work [3, 16, 17], we find that temperature dependence of the resistivity anisotropy in uniaxial strained samples can be well described by a Curie-Weiss functional form above the

strain free T_s and below T^* [see solid lines in Fig. 4(a)-4(d)]. When uniaxial strain is released, the resistivity anisotropy and its appearance temperature T^{**} are dramatically reduced. Nevertheless, it is clearly present above T_s in the tetragonal phase. For strain free samples cooled from high-temperature paramagnetic tetragonal phase, there are no resistivity anisotropy above T_s [see green data points in Fig. 4(a)]. The small resistivity anisotropy below T_N is due to slight imbalance in the twin domain populations.

To understand the observed resistivity anisotropy behavior, we consider a microscopic scenario as shown in Fig. 4(e)-4(h). In the low-temperature uniaxial strained detwinned state, the undoped and underdoped $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ form a single domain homogeneous magnetic ordered state with intrinsic resistivity anisotropy that is weakly electron-doping dependent [Fig. 4(a)-4(d), $\rho_b > \rho_a$]. Upon releasing the uniaxial strain, the sample becomes partially detwinned AF ordered state with reduced resistivity anisotropy [Fig. 4(f)]. On further warming to temperatures above T_N and T_s , these materials exhibit a large lattice distortion across T_N but much less anomaly across T_s [Fig. 3(f) and 3(g)] [7]. These results suggest that the resistivity anisotropy seen in the narrow temperature region above T_s is due to the remnant local lattice distortions arising from the large magnetoelastic coupling across T_N [Fig. 4(g)]. The system finally relaxes to the true homogeneous tetragonal state without resistivity anisotropy at temperatures above T^{**} . Since our neutron Larmor diffraction measurements showed no additional anomaly in lattice parameters and lattice distortion above T_s , we conclude that there is no thermodynamic phase transition at T^* and T^{**} in agreement with recent heat capacity measurements [21]. The resistivity anisotropy seen in the stress free detwinned samples below T^{**} on warming across T_N is then due to local spin nematic correlations and associated lattice distortions arising from the magnetoelastic coupling through the collinear AF state below T_N . The absence of such effect in strain free sample on cooling confirms this scenario and the weakly first order nature of the magnetic transition.

In summary, by using a specially designed in-situ detwinning device, we have discovered the presence of resistivity anisotropy in the tetragonal phase of stress free $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ below T^{**} , a temperature lower than T^* associated with resistivity anisotropy in uniaxial strained sample [13-16]. Our neutron diffraction experiments confirm the partially detwinned state in the stress free sample, thus indicating that the observed resistivity anisotropy arises from local spin nematic correlations and lattice distortions. Furthermore, our neutron Larmor diffraction experiments on lightly electron-doped $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ indicate lattice distortions across T_N and T_s with no evidence of another phase transition above T_s . These results thus establish that resistivity

anisotropy in the tetragonal phase arises from the magnetoelastic coupling associated with static AF order, suggesting the presence of local Ising-nematic spin correlations and lattice distortions in the tetragonal state of electron-doped iron pnictides near T_N .

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Supplementary Materials for: Electronic nematic correlations in the stress free tetragonal state of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$

Sample Information

The $\text{BaFe}_{2-x}\text{Ni}_{1-x}\text{As}_2$ single crystals were grown using self-flux method as described before [1]. The crystal orientation was determined by X-ray Laue machine, and the square shaped samples were cut for Montgomery method resistivity measurements. The samples were annealed at 800 K for 2 days to reduce defects and disorder.

Resistivity measurement: Montgomery method

For sheet like samples, measurement of anisotropic in-plane resistivity can be carried out by Montgomery method [2]. The samples are cut along the a and b axes directions into a squared shape with the c -axis perpendicular to the squared surface. Current is applied through contacts at two adjacent corners of the planar face and the potential is measured at the other two corners at the same plane [SFig 1.(a)]. From the measurements, we can get $R_1 = V_1/I_1$. Similar measurements can be done with the electrical connections rotated 90° with respect to the original setup [SFig1.(b)], which gives $R_2 = V_2/I_2$. This will allow calculations of the resistivity anisotropy.

Results from an anisotropic sample with dimensions l_1, l_2, l_3 , and resistivity ρ_1, ρ_2, ρ_3 can be estimated from an isotropic sample with dimension l'_1, l'_2, l'_3 with the transformation:

$$\rho^3 = \rho_1 \rho_2 \rho_3, \quad (1)$$

and

$$l'_i = l_i(\rho_i/\rho)^{1/2}. \quad (2)$$

Through theoretical calculations, we can get for small l_1/l_2 ratio:

$$\frac{l'_2}{l'_1} \simeq \frac{1}{2} \left[\frac{1}{\pi} \ln \frac{R_2}{R_1} + \sqrt{\left[\frac{1}{\pi} \ln \frac{R_2}{R_1} \right]^2 + 4} \right] \quad (3)$$

and the anisotropic resistivity in the plane can be written as:

$$\rho_1 = A(l'_1/l'_2)R_1 \sinh[\pi l'_2/l'_1] \quad (4)$$

and

$$\rho_2 = B(l'_2/l'_1)R_2 \sinh[\pi l'_1/l'_2]. \quad (5)$$

In the case of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$, A and B are pre-factors that can be normalized as $\rho_1(300 \text{ K}) = \rho_2(300 \text{ K})$. Then ρ_1 and ρ_2 can be determined by resistance measurement

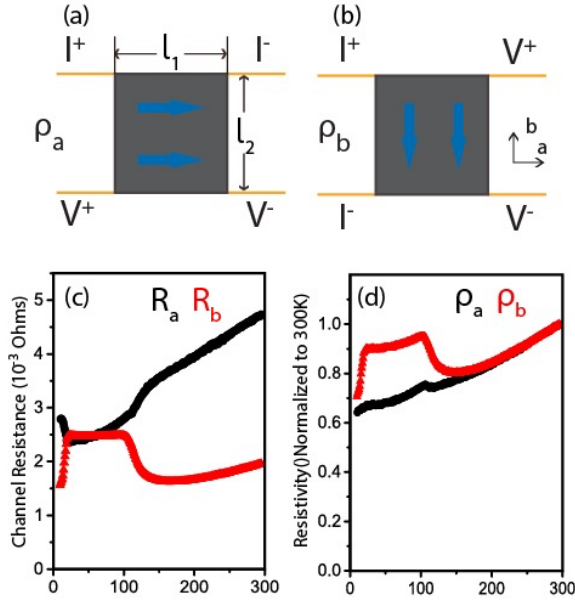


FIG. 5: (a),(b) Schematic illustration of Montgomery method: wire connections and current flow directions. Sample plane dimensions are $l_1 \times l_2$. The blue arrows on the sample are the current direction for each set up. (c) Resistance data of $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ from the two channels on the PPMS resistivity puck. Black and Red indicate the direction of the resistance measured on the channel. (d) Normalized resistivity calculated from (c).

R_1 and R_2 by two different channel on PPMS resistivity puck, as shown in SFig 1.(c) and (d). ρ_1 and ρ_2 derived from Montgomery method is the equivalent resistivity by the mixture of two domains angling 90° with initial resistivity of ρ_{a0} and ρ_{b0} .

Pressure Dependence Measurements of the resistivity anisotropy

The pressure dependence of the in-plane resistivity was studied systematically both as a function of Ni-doping and temperature. The magnitude of the uniaxial pressure applied to the system is determined approximately by the length compression of the spring as shown in Fig. 1(b) of the main text. In SFig. 2(a), we show pressure dependence of the resistivity anisotropy defined as $2(\rho_b - \rho_a)/(\rho_a + \rho_b)$ for zero pressure cooled and pressure cooled case of BaFe_2As_2 . The pressure dependence of the detwinning ratio is also plotted. In the zero pressure cooled case, the sample was cooled down to 10 K with no pressure applied and then temperature was raised to the targeted temperature. In the pressure cooled case, the sample was cooled with maximum pressure (15 MPa) to 10 K, then the pressured is released at 10 K and temperature was raised to the targeted temperature. To the first order approximation, the resistivity anisotropy tracks the

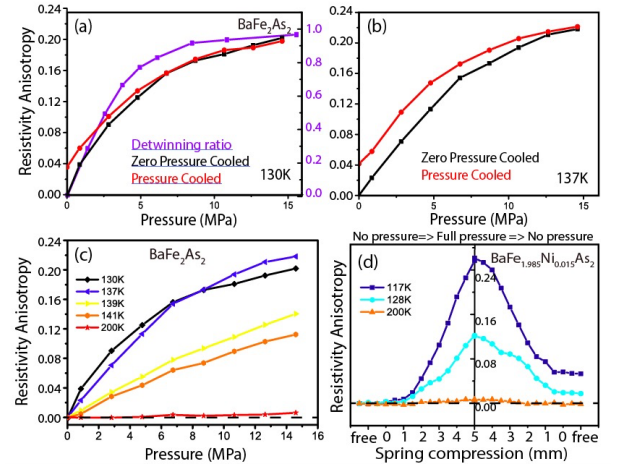


FIG. 6: (a),(b) Resistivity anisotropy of BaFe_2As_2 for the zero pressure cooled (Black) and pressure cooled (Red) sample. Purple line in (a) is the detwinning ratio of BaFe_2As_2 determined by elastic neutron scattering using IN8. (c) pressure dependence of resistivity anisotropy in BaFe_2As_2 (zero pressure cooled) at different temperature. (d) Detwinning effect in $\text{BaFe}_{1.985}\text{Ni}_{0.015}\text{As}_2$ for zero pressure cooled sample at different temperatures.

detwinning ratio of the system before the sample is fully detwinned. SFigure 2(b) shows similar data at 137 K. SFigure 2(c) shows pressure dependence of the resistivity anisotropy across the AF ordering (T_N) and structural (T_s) transitions. For temperatures above T_N and T_s , the pressure and resistivity anisotropy relationship becomes linear and the slope decreases with increasing temperature, consistent with previous work [3]. In SFigure 2(d), we plot the pressure dependence of the resistivity anisotropy at different temperatures below and above T_N and T_s for $\text{BaFe}_{1.985}\text{Ni}_{0.015}\text{As}_2$. Clear hysteresis is seen in the data, suggesting a partially detwinned sample after releasing the pressure. For the pressure released partially detwinned sample, the resistivity anisotropy and the detwinning ratio follow the same trend before T_N , as shown in SFig. 2(a), which suggests the proximate proportionality of resistivity anisotropy with the detwinning ratio η . SFigure 3(a) compares temperature dependence of the detwinning ratio η with that of the resistivity anisotropy. The remarkable similarity in these data again confirms the notion that the reduced resistivity anisotropy in stress free sample is due to reduced detwinning ratio.

Neutron Larmor Diffraction Experiments

Neutron Larmor diffraction is an ideal technique for measuring lattice distortion and expansion, with a resolution better than 10^{-5} for $\Delta d/d$. The resolution is not affected by sample mosaicity or slight sample misalign-

ment, enabling us to keep track of small changes in lattice spacing d and its distortion Δd . Detailed principles of Larmor diffraction technique can be found in references [4–6].

To determine the lattice orthorhombicity induced Bragg peak splitting in the unstrain sample between T_N and T_s , we assume the full-width-half-maximum (FWHM) of the peaks is unchanged across T_s . The result is described in Fig. 3 of the main text.

As discussed in the main text, for a standard second order magnetic phase transition, one would expect that lattice distortion of the system ($\Delta d/d$) remains unchanged across T_N . Since this is clearly not the case for electron underdoped $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$, one can estimate temperature dependence of the lattice correlation length ξ , defined as Fourier transform of the Bragg peak width determined using Larmor diffraction [7]. Assuming that the d -spacing spread follows a Gaussian distribution, the FWHM of its Fourier transform gives the lattice correlation length. For typical triple-axis experiment, the instrument resolution is about 300 Å. Here the resolution is much better as shown in temperature dependence of the lattice correlation length ξ in SFig. 3(b). As a

function of decreasing temperature, the lattice correlation length reduces from 2500 Å at 150 K to 1000 Å at 110 K before becoming 2500 Å again in the AF ordered state.

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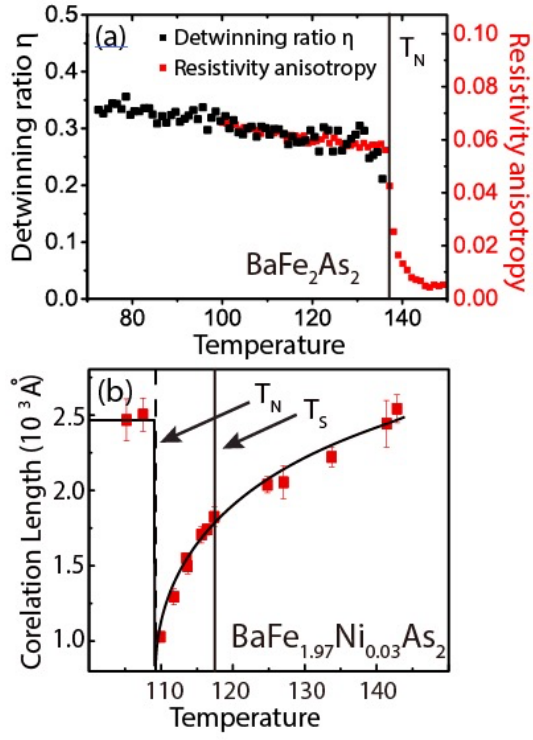


FIG. 7: (a) Temperature dependence of detwinning ratio η (black) and resistivity anisotropy in stress free BaFe_2As_2 . (b) Temperature dependence of the lattice correlation length ξ in $\text{BaFe}_{1.97}\text{Ni}_{0.03}\text{As}_2$ as determined from neutron Larmor diffraction.